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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

FRANCOIS GUGUMUS

APPLICATION NO. 09/899,438

FILED: May 7, 2001

FOR: STABILIZER MIXTURES

Art Unit: 1714

Examiner: M. Medley

Commissioner for Patents

DECLARATION UNDER 37 CFR 1.132

I, François Gugumus, a citizen of France and a resident of 4123 Allschwil, Switzerland, hereby declare:

That I was awarded the degree of a Doctor of Science by the University of Strassbourg, France, in 1965.

That I have been employed by Ciba-Geigy AG, now Ciba Specialty Chemicals Inc., as Chemist in the Additives Division and have been engaged in the field of stabilizers for polymers since 1970.

That I am an inventor of numerous U.S. Patents, for example Nos. 4,148,783; 4,692,486; 4,623,480 and 4,734,449 assigned to Ciba-Geigy Corp., now Ciba Specialty Chemicals Corp..

That I am the author of numerous technical articles relating to the same or closely related field of research, for example:

"Progrès dans la protection des matières plastiques contre le rayonnement UV",  
Kunststoffe-Plastics 22 (1975), 11-19;

"Developments in the UV stabilization of polymers", Developments in Polymer Stabilization, Applied Science Publishers Ltd., London, 1979, 261-308;

"Contribution to polyethylene photooxidation", Angewandte Makromolekulare Chemie 158/159 (1988), 151-176;

"Antioxidantien", Kunststoffe 77 (1987), 1070-1107;

"Lichtschutzmittel", Kunststoffe 77 (1987), 1165-1210;

"The use of accelerated tests in the evaluation of antioxidants and light stabilizers", Developments in Polymer Stabilization, Applied Science Publishers Ltd., London, 1987, 239-289.

That I am the inventor of U.S. Patent Application No. 09/899,438.

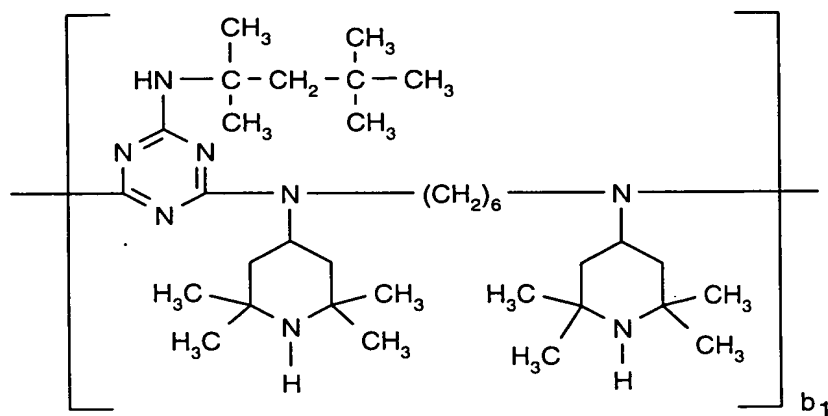
That the experiments described in the following have been made under my supervision and the evaluation of the results has been done by myself.

#### STABILIZERS USED

(The mean degree of polymerization is indicated.)

Compound (B-1-a-1):

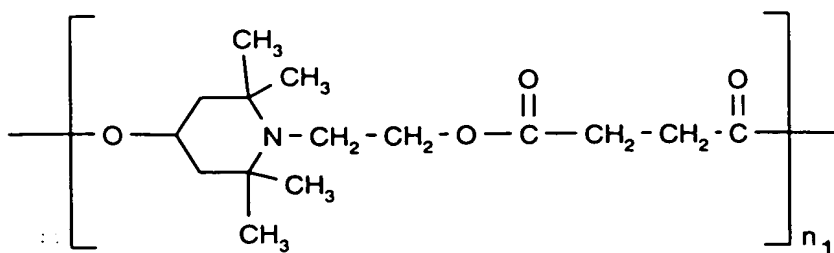
(CHIMASSORB 944 (RTM))



with  $b_1$  being 4.5.

Compound (B-7-a-1):

(TINUVIN 622 (RTM))



with  $n_1$  being 5.1.

## TEST I:

### TEST METHOD

Light stabilization of polypropylene tapes:

100 parts of polypropylene powder (melt flow index: 3.2 g/10 min at 230°C and 2160 g) are blended in a barrel mixer with 0.05 parts of pentaerythrityl tetrakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate], 0.05 parts of tris[2,4-di-tert-butylphenyl] phosphite and the stabilizer system indicated in Table 1. Then, the blend is compounded in an extruder at temperatures of 180°- 220°C. The granules obtained on extrusion and granulation are transformed into films at 220°-260°C in a second extruder equipped with a flat sheet die. The films are cut into ribbons which are drawn to achieve a stretch ratio of 1:6. The tapes obtained with this procedure are finally 50 µm thick and 2.5 mm wide.

The tapes are mounted without tension on sample holders and exposed to natural weathering in Florida (45° South, direct, approximately 145 kLy/year). Periodically, the tensile strength of the exposed tapes is measured. The received energy (in kLy) corresponding to a loss of 50 % ( $E_{50}$ ) of the initial tensile strength or the % retained tensile strength after 320 kLy is a measure for the stabilizing efficiency of the stabilizer system.

The values obtained are summarized in Table 1.

Table 1:

| Sample | Stabilizer System                            | $E_{50}$ (kLy) to 50 % retained tensile strength or<br>% retained tensile strength after 320 kLy |
|--------|--|--|
| 1      | 0.1 % of Mg stearate                         | 58   |
| 2      | 0.1 % of (B-1-a-1) +<br>0.1 % of Mg stearate | 73 %   |

|   |  |      |
|---|--|------|
| 3 | 0.1 % of (B-7-a-1) +<br>0.1 % of Mg stearate                           | 69 % |
| 4 | 0.05 % of (B-1-a-1) +<br>0.05 % of (B-7-a-1) +<br>0.1 % of Mg stearate | 78 % |

Sample 1 does not contain a sterically hindered amine compound but Mg stearate which corresponds to component (II) of the stabilizer mixture according to the present invention. Samples 2 and 3 contain one sterically hindered amine and Mg stearate, while Sample 4 contains the stabilizer mixture according to the present invention, which comprises two different sterically hindered amine compounds (component (I)) and Mg stearate (component (II)).

While Sample 1 has a retained tensile strength of 50 % after an irradiation of only 58 kLy, Sample 2 has a retained tensile strength of 73 % after an irradiation of 320 kLy and Sample 3 has a retained tensile strength of 69 % after an irradiation of 320 kLy, Sample 4 which has been stabilized with a stabilizer mixture according to the present invention has still a retained tensile strength of 78 % after an irradiation of 320 kLy. Such a superiority of Sample 4 is indeed surprising.

## TEST II

Light stabilization of polypropylene homopolymer films:

100 parts of unstabilized polypropylene powder (melt flow index: about 5.5 g/10 min at 230°C and 2160 g) are homogenized at 200°C for 10 min in a Brabender plastograph with 0.05 parts of pentaerythrityl-tetrakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate], 0.05 parts of tris[2,4-di-tert-butylphenyl]phosphite, 0.5 parts of titanium dioxide (rutile) and the amount of light stabilizer and coadditive indicated in Table 1. The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 min at 260°C to a 0.5 mm thick film which is cooled immediately to room temperature in a water-cooled press. Samples of 60 mm x 25 mm are cut out of these 0.5 mm films and are exposed in a WEATHER-OMETER Ci 65 (black panel temperature: 63±2°C, without water-spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer. The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the stabilizing efficiency of the stabilizer mixture. The values obtained are summarized in Table 1.

Table 1:

Light stability of polypropylene films on artificial exposure in a Weather-Ometer Ci 65

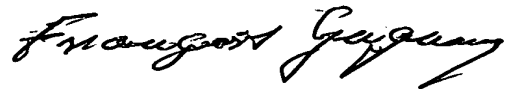
| Stabilizer                                   | Hours to 0.1 carbonyl absorbance*) |                  |
|--|------------------------------------|------------------|
|  | 0.1% Ca stearate                   | 0.1% Mg stearate |
| 0.05% of (B-1-a-1) and<br>0.05% of (B-7-a-1) | 1920                               | 2960             |

\*) High values are desired.

The values above clearly reveal that the stabilizing efficiency of the indicated stabilizer combination is considerably improved in the presence of Mg stearate.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 17<sup>th</sup> day of February 20 04



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François Gugumus